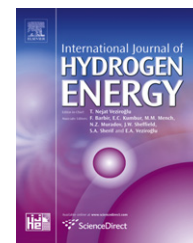


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Short Communication

Thermodynamic analysis of combined unit of biomass gasifier and tar steam reformer for hydrogen production and tar removal

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ABSTRACT

A combined unit of biomass gasifier and tar steam reformer (CGR) was proposed in this study to achieve simultaneous tar removal and increased hydrogen production. Tar steam reforming calculations based on thermodynamic equilibrium were carried out by using Aspen Plus software. Thermodynamic analysis reveals that when selecting appropriate operating conditions, exothermic heat available from the gasifier could sufficiently supply to the heat-demanding units including feed preheaters, steam generator and reformer. The effects of gasification temperature (T_{gs}), reforming temperature (T_{ref}) and steam-to-biomass ratio (S:BM) on percentages of tar removal and improvement of H₂ production were investigated. It was reported that the CGR system can completely remove tar and increase H₂ production (1.6 times) under thermally self-sufficient condition. The increase of H₂ production is mainly via the water–gas shift reaction.

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1. Introduction

Hydrogen energy is considered to be the most attractive energy for the future due to its high energy content and zero greenhouse gas (GHG) emissions. The main technologies for hydrogen production are steam reforming of fossil fuels, and electrolysis and thermolysis of water. Other attractive sources of hydrogen production come from renewable sources, such as bioethanol, biodiesel, biogas and biomass. Although cost of hydrogen production from renewable sources is currently

higher than that from conventional fossil sources, the cost of technology is postulated to reduce in the near future [1,2]. Gasification technology has been applied to convert biomass into fuel gas, whose main components are methane, hydrogen and carbon monoxide. Fuel gas can be further used for production of thermal heat by direct combustion, electricity by power generator or fuel cells, and synthesis fuels by Fischer–Tropsch process (FT). Biomass gasification has been investigated in both experimental and mathematical modeling studies. A number of efforts have been devoted to improve

Abbreviations: CGR, combined unit of biomass gasification and tar reformer; conv, conventional system; MSR, methane steam reforming; TSR, tar steam reforming; WGSR, water–gas shift reaction.

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technologies of biomass gasification, for examples, by adding steam [3,4] or CO₂ [5–9] with air in feed, by integration with CO₂ capture [3,4,10,11] and by using different catalysts [5–9,12].

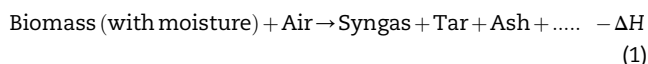
The major problem of biomass gasification is tar (high molecular weight hydrocarbon compounds) [12–18]. Tar in fuel gas from gasification can condense at low temperature, leading to clogging and/or blocking of pipe lines, valves, filters, engines and fuel cell stacks. Tar can be removed by a physical process (e.g. liquid scrubber, carbon adsorption and electrostatic precipitator) [13,14]. Increasing gasification temperature is another promising approach to reduce tar formation [15–18]. It is also possible to install a catalytic tar reformer for tar removal. There are a number of researches focusing on development of suitable catalysts and determination of appropriate operating condition for tar reforming [12,19–29]. Fuel gas compositions from gasification of Indian biomass were determined based on four atomic balances (C, H, O and N) and equilibrium reactions. The gross calorific value (GCV) was used as the main criteria for engine application. GCV was found to increase with increasing temperature. Gordillo and Annamalai [12] studied dairy biomass gasification with air and steam. The results showed that CH₄ and H₂ increase with increasing steam to fuel ratio (S:F). Thermodynamic performance of gasification of oil palm shell was investigated by methane steam reforming and water–gas shift reaction for producing hydrogen [30]. Thermodynamic parameters are determined for design, optimization and modification of hydrogen production from biomass. Biomass gasification was studied under supercritical water condition by using solar energy as an energy source [31].

This work was aimed at determining the efficiency of the proposed combined unit of biomass gasifier and tar steam reformer (CGR) for simultaneous tar removal and improved hydrogen production by using thermodynamic analysis. The effects of various operating parameters such as temperature and steam:biomass ratio on the tar removal, product gas composition and thermal energy are investigated. Particular focus is on the conditions which do not demand additional energy from external sources.

2. Theory

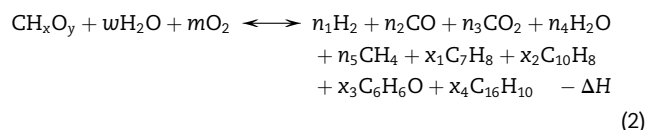
2.1. Biomass gasification

Gasification is a process to convert carbonaceous materials such as coal, petroleum, biofuel, or biomass into carbon monoxide and hydrogen. Carbonaceous materials react with controlled amount of oxygen and/or steam (Eq. (1)) at high temperatures, resulting in a gas mixture called synthesis gas or syngas.



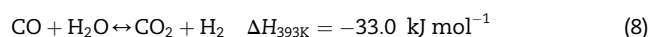
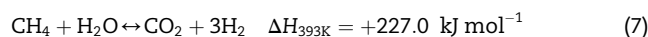
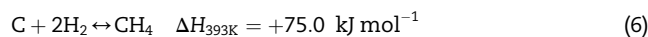
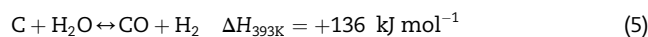
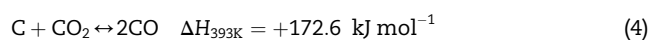
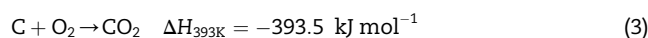
The tar compositions were investigated and reported by Brage et al. [32] for wood gasification at various operating temperatures (973–1173 K). Singh et al. [15] used toluene (C₇H₈) to represent all the one-ring compounds, naphthalene (C₁₀H₈) to represent two-ring compounds, phenol (C₆H₆O) to

represent phenolic and other heterocyclic compounds and pyrene (C₁₆H₁₀) to represent three-rings and higher compounds. Tar components used for calculations in this study were based on the representative tar components from biomass gasification whose compositions varied with gasification temperature. Table 1 summarizes the properties of wood chip (East-India walnut), which is represented by CH_{1.79}O_{0.83} (neglecting small extent of N of about 1.3 × 10^{−3}). An overall reaction of biomass gasification is shown in Eq. (2),



where w = stoichiometric of biomass moisture, m = stoichiometric of oxygen, n_i = stoichiometric of reaction, x_i = mole of representative of tar.

The reactions involved in gasification (Eqs. (3)–(8)) were reported by Wang and Kinoshita [31].



In this study, the reactions in the gasifier were assumed at their thermodynamic equilibrium. The calculations were based on Gibbs Free energy minimization and atomic balances. The equations were solved using Newton–Raphson method in MATLAB program. The model results from this study were validated with those from experimental results [33] and mathematical models [34,35] of downdraft biomass gasification (Table 2). These results may not be exactly at the same conditions (biomass types, humidity and oxygen-to-biomass ratio). The results show that CO and CO₂ are close to the experimental data [33] and the model data [34,35]. On the other hand, lower contents of H₂ and CH₄ are obtained probably because the model in this study was calculated

Table 1 – Summary of wood properties.

| | |
|---------------------------|--------------------------|
| <i>Proximate analysis</i> | |
| Volatile matter | 74.76 wt% |
| Fixed carbon | 13.66 wt% |
| Ash | 4.38 wt% |
| Moisture content | 5.41 wt% |
| High heating value | 18.87 MJkg ^{−1} |
| <i>Ultimate analysis</i> | |
| Carbon, C | 44.24 wt% |
| Hydrogen, H | 6.548 wt% |
| Nitrogen, N | 0.071 wt% |
| Oxygen, O | 49.141 wt% |

Table 2 – Comparison of results with literatures [mol% (dry basis)].

| | CO ₂ | CO | H ₂ | CH ₄ |
|---|-----------------|-------|----------------|-----------------|
| Experiment [33] | 12.17 | 18.24 | 27.65 | 2.06 |
| Model [34] | 12.85 | 22.72 | 27.91 | 1.58 |
| Model [35] | 18.72 | 10.46 | 30.90 | 5.93 |
| This study (at $T_{gs} = 873$ K, moisture content = 5% and $O_2:BM = 0.5$) $CH_{1.451}O_{0.697}$ | 12.31 | 16.52 | 13.31 | 0.02 |

based on compositions of representative tar and the operating condition was not the same.

2.2. Tar removal

Tar removal is mostly carried out by cooling down the fuel gas to a temperature lower than its tar dew point temperature (usually greater than 573 K). Water is used to cool down the fuel gas temperature and to separate tar from gas phase into liquid phase (Fig. 1). In this study, the tar removal system was a tar steam reformer whose products were calculated using Aspen Plus Program with RGibb module (all possible reaction products were specified). Thermodynamic properties were calculated by using PENG-ROB thermodynamic property estimation method. The operation was assumed to take place under adiabatic condition. The results of product gas temperature and tar composition were reported.

2.3. Tar steam reforming

Representative tars composed of toluene, naphthalene, phenol and pyrene. The reactions involved are tar steam reforming (TSR), methane steam reforming (MSR) and water–gas shift reaction (WGSR) as shown in Eqs. (9)–(15).

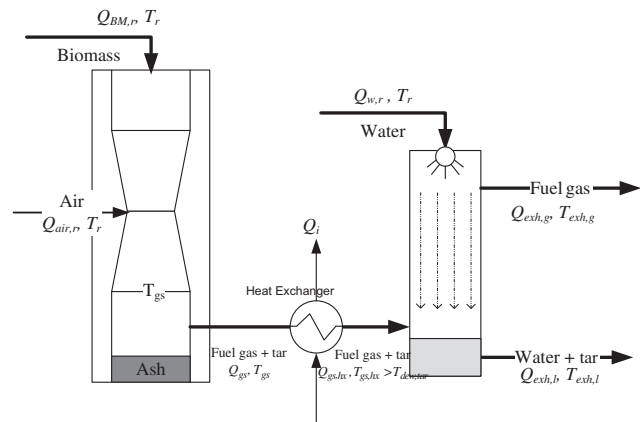
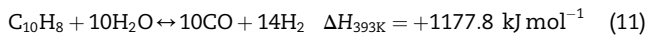
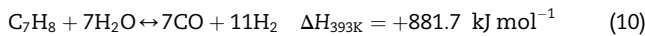
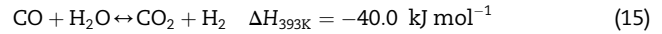
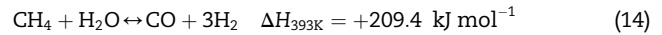
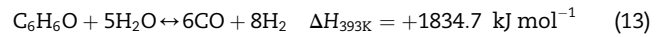
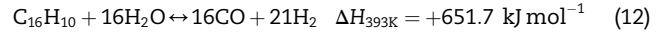


Fig. 1 – Conventional biomass gasification with tar removal unit.



The proposed combined unit of biomass gasifier and tar steam reformer (CGR) is shown in Fig. 2. The overall gasification reactions are exothermic while the steam reforming reactions are endothermic. Heat energy is also needed at a steam generator and heating units. From Fig. 2, exothermic energy from the gasification section could supply to the heat-demanding units including feed preheaters, steam generator and the reformer at various operating temperature. The thermal energy involved at different units of CGR can be determined by using conventional energy balances. Eq. (16) is the overall thermal energy (Q_{tot}) of the proposed system. Q_{gs} represents heat energy of gasification (Eq. (17)) including heat energy involved in gasification and in preheating of biomass (Eq. (19)) and air (Eq. (20)). Q_{st} represents heat energy for the steam generator (Eq. (21)) while Q_{ref} represents heat energy of the tar reformer (Eq. (22)). Generally, the energy from gasification is highly exothermic; therefore, it is possible to supply to other heat-demanding units in the CGR system. When operating conditions of the system (e.g. operating temperatures and steam-to-biomass molar ratio) are carefully selected, the value of Q_{tot} becomes less than zero, indicating that the system can be operated without demanding additional energy for external source.

$$Q_{\text{tot}} = \sum Q = Q_{\text{gs}} + Q_{\text{st}} + Q_{\text{ref}} \quad (16)$$

where

$$Q_{\text{gs}} = Q_{\text{rxn,gs}} + Q_{\text{air,gs}} + Q_{\text{bm,gs}} \quad (17)$$

$$Q_{\text{rxn,gs}} = \left(\sum_i^n \Delta H_{\text{rxn},i} \right) \quad (18)$$

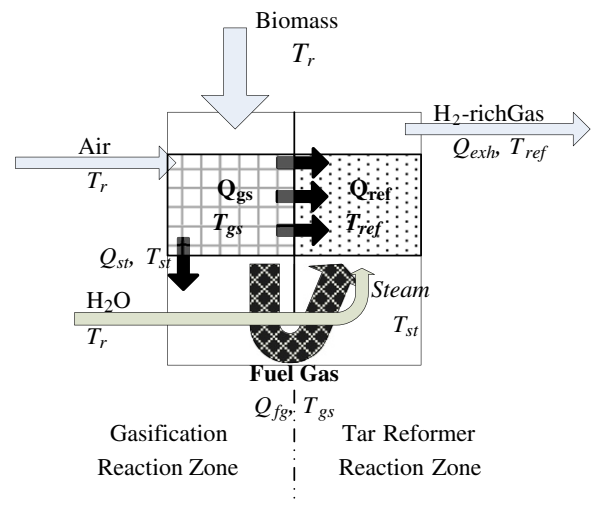


Fig. 2 – Scheme of combined unit of biomass gasification and tar steam reformer (CGR).

$$Q_{bm,gs} = (m \cdot c_p \cdot \Delta T)_{bm} \quad (19)$$

$$Q_{air,gs} = (m \cdot c_p \cdot \Delta T)_{air} \quad (20)$$

$$Q_{st} = \left(m \cdot \left((c_p \cdot \Delta T)_{liq} + L + (c_p \cdot \Delta T)_{gas} \right) \right)_{H_2O} \quad (21)$$

$$Q_{ref} = \left(\sum_j^n H_{TSR,j} \right) + H_{MSR} + H_{WGR} \quad (22)$$

In this study, H_2 production improvement (Eq. (23)) is the key performance indicator used for comparison between the proposed CGR and the conventional gasification process.

$$H_{2,imp} = H_{2,CGR} / H_{2,conv} \quad (23)$$

3. Results and discussion

Standard operating condition of biomass gasification for this study is shown in Table 3. Gasification reaction products of biomass [24] were calculated at different temperatures (973–1173 K). The results of fuel gas compositions (solid line) and thermal energy (dash line) are shown in Fig. 3. When increasing gasification temperature from 973 to 1173 K, hydrogen slightly drops from 14.37 to 13.60 mol% (dry basis), carbon dioxide decreases from 16.32 to 13.55 mol% (dry basis), carbon monoxide rises from 10.52 to 15.07 mol% (dry basis) and methane decreases from 0.02 to 0.0001 mol% (dry basis). In addition, the total thermal energy of gasification (Q_{gs}), calculated by summation of biomass gasification reaction ($Q_{rxn,gs}$), air heating ($Q_{air,gs}$), and biomass heating ($Q_{bm,gs}$), changes from -0.99 to -0.45 MJ h^{-1} . $Q_{rxn,gs}$ is relatively constant (-2.98 to -3.05 MJ h^{-1}), $Q_{air,gs}$ increases from 1.62 to 2.14 MJ h^{-1} and $Q_{bm,gs}$ increases from 0.36 to 0.47 MJ h^{-1} . In summary, the increasing gasification temperature (T_{gs}) results in increasing CO, but decreasing H_2 and Q_{gs} . Fig. 4, shows the fraction of tar compositions at different operating conditions, consisting of C_6H_6O , C_7H_8 , $C_{10}H_8$ and $C_{16}H_{10}$ which are used in this study [21]. The tar composition is reduced from about 0.42 to 0.17 mol% (dry basis) when increasing operating temperature from 973 to 1173 K. At lower operating temperature (973 K), phenol (C_6H_6O) is the major tar component (about 0.25 mol% (dry basis)). At higher operating temperature (1173 K), C_6H_6O is reduced to 0.056 mol% (dry basis). C_7H_8 is decreased from 0.085 to 0.028 mol% (dry basis) and $C_{10}H_8$ slightly rises from 0.061 to 0.079 mol% (dry basis), when increasing operating temperature for 973 to 1173 K. $C_{16}H_{10}$ is close to zero (about $10^{-3} \text{ mol h}^{-1}$). From Fig. 4, it is clear that tar can be reduced by increasing gasification temperature.

Table 3 – Standard operating condition of biomass gasification.

| Parameter | Value |
|---|-------------------------|
| Biomass | 1 [kg h ⁻¹] |
| Gasification temperature (T_{gs}) | 973–1173 [K] |
| Oxygen-to-biomass mole ratio ($O_2:BM$) | 1:2 [–] |
| Steam temperature (T_{st}) | 673–973 [K] |

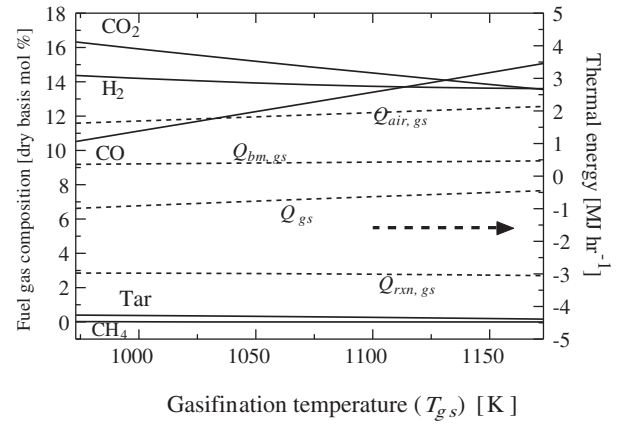


Fig. 3 – Fuel gas compositions (dry basis) and thermal energy at various gasification temperatures ($O_2:BM = 1:2$ and moisture content = 5.41%).

Considering the conventional tar removal unit carried out by water spraying to cool down the gas temperature to a value lower than its tar dew point temperature. Fig. 5 shows the effects of water-to-biomass molar ratio (W:BM) and gasification temperature (T_{gs}) on fuel gas temperature ($T_{out,tr}$) and tar removal [%]. The calculations assumed that the tar removal unit was operated under adiabatic condition and phase equilibrium; therefore, the fuel gas temperature ($T_{out,tr}$) as well as the product gas composition could be determined. It was found that the fuel gas temperature ($T_{out,tr}$) decreases with increasing water-to-biomass ratio (W:BM) and decreasing gasification temperature (T_{gs}). Tar removal significantly increases when increasing water at low range of W:BM ratio and levels off at higher values. However, there is no monotonic relationship between tar removal and gasification temperature (T_{gs}). Maximum tar removal appears at T_{gs} of 1073 K. This is due to the differences in tar compositions at different gasification temperature (T_{gs}) (as shown in Fig. 4) which also have different solubility in water. It is noted that W:BM ratio as high as 15–24 is required to remove 99% of tar from the gasification products. The resulting fuel gas temperatures ($T_{out,tr}$) are lower than 350 K ($T_{out,tr} = 340.9, 342.5, 344.5, 345.9$ and 347.5 K for $T_{gs} = 973, 1023, 1073, 1123$ and 1173 K ,

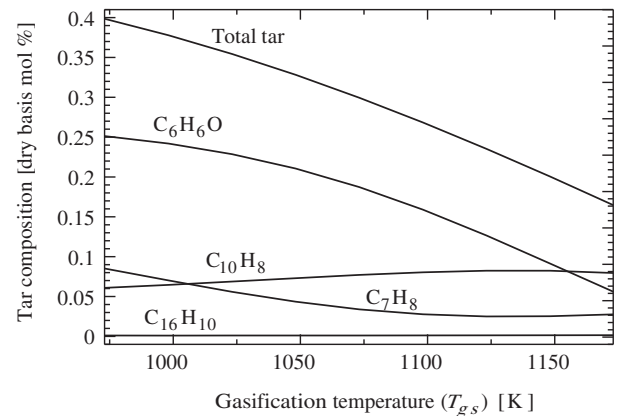


Fig. 4 – Tar compositions from gasification at different temperatures ($O_2:BM = 1:2$ and moisture content = 5.41%).

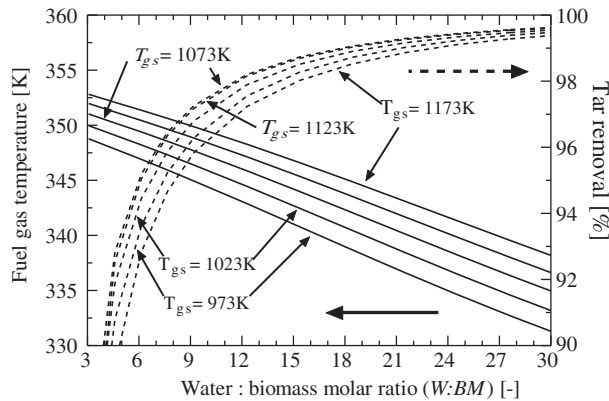


Fig. 5 – Effect of water-to-biomass molar ratio on tar removal (%) and output fuel gas temperature ($T_{out,tr}$).

respectively). From these results, it is clear that the conventional tar removal is an inefficient system in terms of high environmental impact (consuming high water resources) and poor thermal efficiency (temperature out ~ 350 K). Consequently, this study proposed the system which offers reduced environmental impact and improved thermal efficiency of the biomass gasification system. The calculation results of the combined unit of biomass gasification and tar steam reformer (CGR) are shown in Fig. 6. Fuel gas from the gasification zone reacts with steam from the steam generator (supplied heat energy from fuel gas) in the tar steam reformer zone. The values of the steam:biomass molar ratio (S:BM) were varied between 0–4 [–]. Fig. 6 shows gas compositions from the tar steam reformer from the case with a gasification temperature (T_{gs}) of 973 K. The gas compositions between fuel gas input (from Fig. 3) and fuel gas output (solid line) were compared. The maximum H_2 of 24.7 mol% (dry basis) appears at the S:BM ratio of 3.5. At this condition, the H_2 production improvement, as high as 1.72 times of the value from the case of single gasification, was achieved, because additional H_2 was generated from the reactions of steam with both representative tar and CO (WGSR). It is found that the representative tars were completely converted to syngas (H_2 and CO_2). These

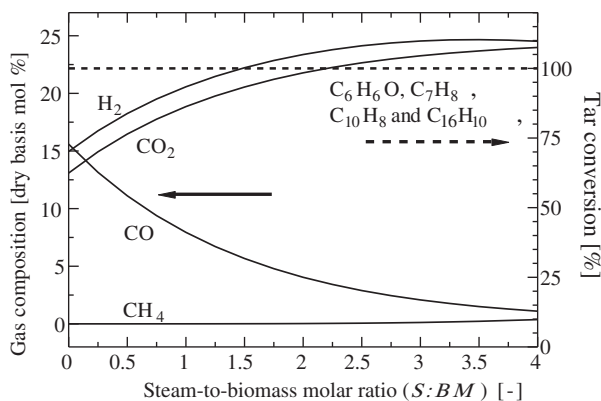


Fig. 6 – Effect of steam-to-biomass molar ratio (S:BM) on product gas compositions from tar reformer (solid line) and tar conversion (dash line) ($T_{gs} = 973$ K).

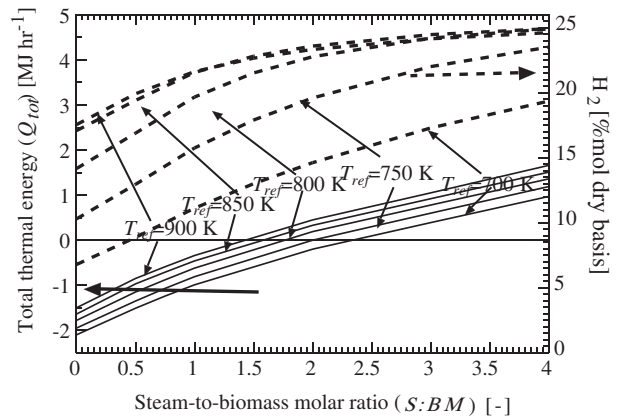


Fig. 7 – Effect of reforming temperature (T_{ref}) and steam-to-biomass molar ratio (S:BM) on hydrogen production and total thermal energy (Q_{tot}) ($T_{gs} = 973$ K).

results show the possibility of utilizing only internal heat energy of the gasification for reduction of tar. However, it should be noted that the major increase of H_2 was dominated by water–gas shift reaction (WGSR), from the observation of the reduced CO. At lower values of the S:BM ratio (< 0.57), the CO composition is higher than that of the fuel gas input (10.3 mol% (dry basis)). The results indicate that tar steam reforming is an effective reaction to remove tar, and water–gas shift reaction is an improvement in the system, since CO is reduced and H_2 is increased when increasing S:BM ratio ($0.57 < \text{S:BM} < 3.5$). At higher S:BM (> 3.5), production H_2 slightly drops while CH_4 slightly increases, indicating that the methanation reaction becomes important.

Thermal energy balance and H_2 production of CGR system operated at various values of S:BM ratio and reforming temperature (T_{ref}) at $T_{gs} = 973$ K are shown in Fig. 7. Total thermal energy (Q_{tot}) of CGR system illustrates possibility of utilizing energy of gasification for tar reforming in the CGR. When total thermal energy is higher than zero, the CGR system requires external thermal energy for supplying to the system. The suitable S:BM ratios for thermally self-sufficient conditions of

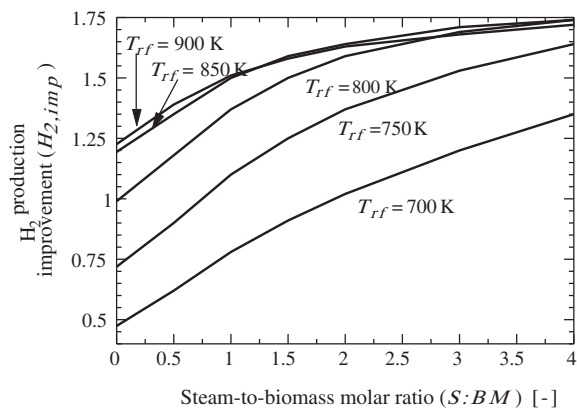


Fig. 8 – H_2 improvement of integrated tar steam reformer on biomass gasification compared with conventional biomass gasification ($T_{gs} = 973$ K and W:BM = 15).

Table 4 – Maximum H₂ production improvement (H_{2,imp}) (at T_{gs} = 973 K and W:BM = 15).

| Reforming temperature (T _{ref}) [K] | Values | |
|---|----------|------------------------|
| | S:BM [–] | H _{2,imp} [–] |
| 700 | 2.28 | 1.07 |
| 750 | 2.05 | 1.37 |
| 800 | 1.82 | 1.55 |
| 850 | 1.58 | 1.60 |
| 900 | 1.38 | 1.57 |

various reforming temperatures (T_{ref} = 700, 750, 800, 850 and 900 K) are 2.3, 2.0, 1.8, 1.6 and 1.4, respectively. H₂ production increases when increasing S:BM ratio; however the condition of thermally self-sufficient system should be of concern. Fig. 8 shows H₂ production improvement (defined as Eq. (23)) of fuel gas compared with the conventional biomass gasification, at a tar removal of 99%. It illustrates the contour of H₂ production improvement at various reforming temperatures (T_{ref}) and steam: biomass ratios (S:BM). The improvement of H₂ production as high as 1.6 times over the conventional biomass gasification could be achieved. Table 4 summarizes the maximum improvement of H₂ production and its corresponding S:BM ratio at different values of reforming temperature for the thermally self-sufficient system.

4. Conclusion

The combined unit of biomass gasifier and tar steam reformer (CGR) has been proven in this study to be an efficient technology for hydrogen production. The unit allows simultaneous tar removal and improved hydrogen production. The calculations were carried out using Aspen Plus program. By selecting suitable operating parameters (gasification temperature (T_{gs}), reforming temperature (T_{ref}) and steam-to-biomass ratio (S:BM)), the exothermic heat from gasification can sufficiently supply to the other heat-demanding units including the steam reformer. It is demonstrated that the improvement of H₂ production as high as 1.6 could be achieved in the proposed unit.

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Nomenclature

c_p : heat capacity, kJ (mol K)^{-1}
 H_2 : hydrogen production, mol h^{-1}
 $H_{2,imp}$: hydrogen production improvement, –
 L : latent heat, kJ mol^{-1}
 Q_{fg} : heat of fuel gas, MJ h^{-1}
 Q_{gs} : heat of gasification, MJ h^{-1}
 Q_{out} : heat of exhaust gas, MJ h^{-1}
 Q_{tot} : total thermal energy, MJ h^{-1}
 $S:BM$: steam-to-biomass mole ratio, –
 T_{fg} : fuel gas temperature, K
 T_{gs} : gasifier temperature, K
 $T_{out,tr}$: output tar reformer temperature, K
 T_r : room temperature (293 K), K
 T_{ref} : tar steam reforming temperature, K
 $W:BM$: water-to-biomass mole ratio, –